Assessment of pollutant origin in road runoff sediments

Étude de la provenance des polluants dans les sédiments des eaux de ruissellement routières

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ABSTRACT

Treatment from road runoff generates considerable amounts of polluted sediments. Several previous studies have assessed the quality and generation rate of road runoff sediments in different types of stormwater treatment facilities. In those studies the origin of the pollutants, e.g. a roadside deposit, has been discussed, but to date, it has not been possible to quantify pollutants generated directly by the road environment or atmospheric deposition. In this study, the origin of sediments generated from highly trafficked roads and road tunnels, wash water from road tunnels and road surfacing materials was assessed. The Cr and Ni present in asphalt material could have contributed to concentrations found in sediments from road runoff in the watersheds studied. For the PAH found in runoff sediments, traffic was identified as a significant source. The Zn and Pb in the runoff sediments probably have additional sources besides traffic and the traffic environment. From the results, it could also be concluded that PAH, Co, Cu, Pb and Zn concentrations in runoff sediments do not originate from asphalt to any significant extent.

RÉSUMÉ

Le traitement des eaux pluviales routières génère des quantités considérables de sédiments pollués. Plusieurs études ont été menées pour évaluer la qualité et le taux de génération des sédiments des eaux pluviales routières dans différents types d'installations de traitement des eaux pluviales. Des recherches antérieures ont déjà étudié l'origine des polluants, par exemple celui des dépôts de la route. Cependant, il n'a pas encore été possible d'estimer les polluants générés directement par l'environnement routier ou par les dépôts atmosphériques. Dans cette étude, l'origine des polluants a été évaluée en examinant des sédiments provenant de routes à fort trafic, de tunnels routiers, de l'eau de lavage de tunnels routiers et enfin, de sédiments de chaussée de route. Cr et Ni relevés dans l'asphalte pourrait ainsi contribuer aux concentrations trouvées dans les sédiments des eaux pluviales routières dans les bassins hydrographiques étudiés. Le trafic est probablement une cause importante des hydrocarbures aromatiques polycycliques (HAP) dans les sédiments des eaux pluviales routières évaluées. Zn et Pb dans les sédiments évalués devraient également avoir d'autres sources que le trafic et l'environnement routier. De cette étude on pourrait aussi conclure, mais pas de manière importante, que les concentrations de HAP, Co, Cu, Pb et Zn dans les sédiments des eaux pluviales routières proviennent de l'asphalte.

KEYWORDS

Abrasion test, Heavy metals, Organic pollutants, Road pavement, Tunnel wash

1 INTRODUCTION

Road-deposited pollutants originate from a complex mixture of sources. Potential sources include atmospheric deposition and emissions related to traffic (Gunawardana et al., 2012). Many metals are associated with motor vehicles and present in tyres, brake pads and exhaust emissions. Several studies have attempted to assess the contribution of traffic-related sources to deposited pollutants. Research results confirm that heavy metals such as Zn, Cu, Pb, Ni and Cr are generated from brake and tyre wear (Gunawardana et al., 2012; McKenzie et al., 2009). Brake wear gives high concentrations of Cu, while a significant source of Zn is associated with tyre wear (Apeagyei et al., 2011). Davis et al. (2001) reported that brake emissions from vehicles contributed to 47% of the total Cu loading in urban runoff, while 25% of total Zn loading in urban runoff was associated with tyre wear. Comparisons of brake composition showed that the Cu concentration was 50-fold higher than the Cr concentration and 100-fold higher than the Ni concentration. Corresponding comparisons of tyre composition showed that the Zn concentration was 6-fold higher than the Cr concentration and 15-fold higher than the Ni concentration (McKenzie et al., 2009). Kreider et al. (2010) studied the interaction of tyres and road surfaces and reported that although tyre wear is a major contributor of the Zn in road dust, asphalt abrasion and non-traffic related sources may also contribute. The same study reported that the most significant contribution of polycyclic aromatic hydrocarbons (PAH) was from non-tyre sources.

In many countries asphalt is the most common road surfacing material. The traffic load usually specifies the composition of the asphalt mixture used, but asphalt generally contains about 95% stone material and 5% bitumen. The particles from the stone material in the asphalt have been found to have a high capacity for adsorbing metal ions (Lindgren, 1996). Lindgren (1998) analysed bitumen and found concentrations for the sum of 16 PAH to be between 26 and 56 mg/kg, so the bitumen in the asphalt material is also a potential source of PAH. Other sources of PAH in the traffic environment include motor oils (Latimer et al, 1990) and car exhausts (Takada et al., 1991). In winter, the stone material in the asphalt is considered to contribute to the metal pollutant load in road runoff (Lindgren, 1996). Meland et al. (2010b) indicated that the heavy metal pollutants found in tunnel wash water and to some extent also PAH originated from asphalt wear.

Particulate material in road runoff contains metal and organic pollutants (e.g. Hallberg et al., 2006; Boxall and Maltby, 1997; Sansalone and Buchberger, 1997). During winter, the particle generation from road pavements in cold climate regions is accentuated by measures such as application of friction sand and salt and the use of studded tires (VTI, 2005; Kupiainen and Tervahattu, 2005; Jacobson and Hornwall, 1999). This combined with high winter runoff raises pollutant loads (e.g. UK Environmental Agency, 2003; Glenn and Sansalone, 2002; Legret and Pagotto, 1999). Hallberg et al. (2006) showed that metals in highway runoff from asphalt paving have a strong correlation to the particulate matter in winter.

Today there is an increased use of road tunnels (e.g. Meland et al., 2010a,b; Trafikverket, 2009). The tunnel environment is more directly exposed to vehicle pollutants than roads in the open (Paruch and Roseth 2008) as well as the wear of particles from the paving material. During tunnel maintenance, the wash water runoff produced has been found to contain elevated pollutant loads (Meland et al., 2010b), particularly zinc (e.g. Andersen and Vethe, 1994). Paruch and Roseth (2008) noted that most of pollution components in tunnel wash water are attached to particles.

In this study, sediment generated by asphalt material designed for major roads with an annual average daily traffic (AADT) exceeding 7,000 vehicles per lane was studied using a standardised laboratory abrasion test. The results from the abrasion test were compared with field data from two separate watersheds and sediments selectively generated during a tunnel wash.

2 STUDY AREAS

2.1 Fredhäll watershed

The Fredshäll watershed and its runoff quality have been described earlier by Hallberg (2007). The watershed is a section of the European motorway system E4 in the central area of Stockholm, Sweden, and the AADT is 120,000. The runoff is collected in a sedimentation basin and sedimentation is carried out batch-wise, with a minimum retention time of 36 hours. The reduction in suspended solids after the batch-wise sedimentation is typically over 90% (Trafikverket, 2007a).

2.2 Södra Länken watershed

Södra Länken is a tunnel system in the south of Stockholm that comprises a total of 18 km of road tunnel (Trafikverket, 2007b). The traffic in the tunnel is separated in dual tunnels, each with two lanes. The AADT is 100,000. The road surfacing material is asphalt and the tunnel walls are sprayed with concrete (“shotcrete”). The tunnel is regularly washed and the tunnel wash water is discharged to sedimentation basins where it is treated batch-wise for 36 hours. The same sedimentation basins are used for treating the road runoff. The reduction in particulate material after the batch-wise treatment is well over 90%. The road runoff is collected from a catchment area of 2.5 ha.

2.3 The Törnskogs tunnel watershed

The Törnskogs tunnel environment is very similar to that at Södra Länken (e.g. shotcrete on the walls of the tunnel). The Törnskogs tunnel is located approximately 20 km north of Stockholm, is 2.1 km long and consists of two tunnel tubes, one with two lanes for each direction. The AADT is 20,000-25,000. The ventilation system in the Törnskogs tunnel is a combination of natural and forced ventilation. There are no ventilation shafts. All wash water generated during tunnel washing is collected by the tunnel’s drainage system and transported by gravity to the treatment plant. The combined tunnel wash water and stormwater drainage system is separated from the drainage system for groundwater.

3 MATERIALS AND METHODS

3.1 Abrasion test procedure

The abrasion test was carried out using the Prall method (SS-EN 12697-16, Method A). In the standard method, the same water is circulated to and from a cooling basin to keep the temperature during abrasion at 5 °C. For the present study, the cooling water loop was replaced and connected to the pressurised distribution net for drinking water and the discharge after abrasion was diverted to a sedimentation column (Fig. 1).

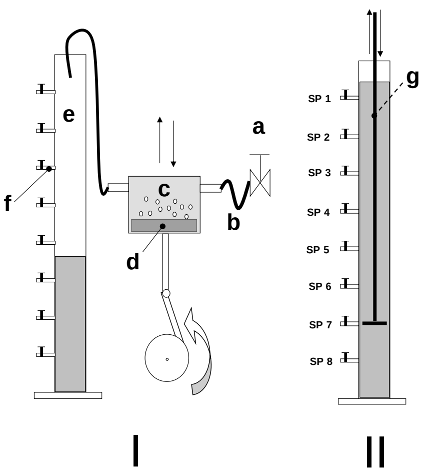

Figure 1 Prall test procedure: I – Specimen abrasion, II – Homogenisation before sampling, a = control valve water flow, b = flexible plastic hose, c = steel spheres, d= asphalt specimen, e = plastic column, f = sampling port (SP), g = stirring rod for homogenisation of the volume.

Drinking water has very low concentrations of the parameters studied here and these were assumed not to influence the results. The content of pollutants (mg/kg) was calculated by analysis of total suspended solids (TSS), total concentrations of the pollutants and dissolved concentrations. The samples were drawn from the sedimentation columns after complete homogenisation of the water column.

A description of the asphalt specimens tested is given in Table 1. Specimen A was sampled from production batches for the surfacing material that would be replacing the surfacing on the road section from which specimen B was collected. The specimens used in the abrasion test were stamped according to standard method SS-EN 12697-30. Asphalt specimen B was removed from a road which from part of the Fredhäll catchment area, after four years of use. This asphalt composition is also used in the tunnel sections in Södra Länken and in the Törnskogs tunnel.

Table 1 Description of asphalt mixtures A and B. Suffixes 1 and 2 refer to the individual samples of each asphalt type.

Specimen	Type	Bitumen wt-%	Origin	Stone material
A1, A2	ABS 16 70/100	6.1	Production batch	Porphyry, quartzite
B1, B2	ABS 16 70/100	5.5	Road surface after four (4) years of use	Porphyry, quartzite

3.2 Fredhäll – Sediment sampling

The sediment was collected after the basin had been completely emptied. A total of three samples were collected from the inlet to the outlet. Each of the three samples in turn was a combined sample of three sampling points in the basin. Sampling was carried out on two occasions, after 403 days and 350 days, during the period January 2004 to February 2006.

3.3 Södra Länken – Sediment sampling

The sediment was collected in 900 mm high sediment traps with collection tube diameter of 75 mm. The 13 sediment traps used were evenly distributed from the inlets to the outlets of two parallel sedimentation basins. Sampling was carried out on three occasions, after 329 days, 174 days, and 183 days, during the period December 2004 October 2006.

3.4 The Törnskog tunnel – Sediment sampling

The wash water was sampled with a submersible pump from the inlet channel to six sampling vessels, each with a total volume of 200 L. The sampling pump was placed after a mechanical screen (3 mm). The collection of tunnel wash water and sedimentation is described in detail by Byman (2012). Sediment samples were collected after sedimentation using a flocculent and decanting of the supernatant from each of the sedimentation vessels. The initial TSS concentration before sedimentation ranged from 1240 to 9690 mg/L. The reduction in TSS after sedimentation was over 98%. The water used for washing was drinking water, and addition of the studied pollutants from the drinking water was assumed to be negligible.

3.5 Chemical analysis of metals and PAH

All analyses were carried out by an accredited laboratory (SWEDAC ISO/IEC 17025, Reg. number 1087). Metals were analysed by adding two mL HNO_3 (suprapur) to 20 mL sample water, which was then digested in a microwave oven. Analysis of the metals was performed using EPA method (modified) 200.7 (ICP-SFMS). A 0.45 µm filter was used for the filtered samples. Analysis of TSS carried out according to SS-EN 872. The sediment was dissolved in a microwave oven in sealed Teflon containers using 1:1 sulphuric acid and water and analysis of metals was carried out according to EPA methods 200.7 (ICP-AES, Inductively Coupled Plasma-Atomic Emission Spectrometry) and 200.8 (ICP-MS, Inductively Coupled Plasma-Mass Spectrometry). PAH analysis is carried out by GC-MS (Gas Chromatography-MS). The laboratory responsible for the analyses also provided all flasks and vessels for sampling.

4 RESULTS AND DISCUSSION

All the study sites were in the same geographical area and the road surfacing was all of the same basic recipe. The sediment in Fredhäll derived from road runoff during rain and snowmelt, while that from Södra Länken was influenced by tunnel wash water but originated mainly from road runoff. The sediment collected in the Törnskog tunnel was pre-dominantly generated during washing. The sediment generated during the abrasion test provided baseline values for the composition of particles from the road surfacing material.

The PAH concentration in the sediments from the abrasion tests was at least 9-fold lower than the PAH concentrations in the study areas (Fig. 2, Tables 2-5).

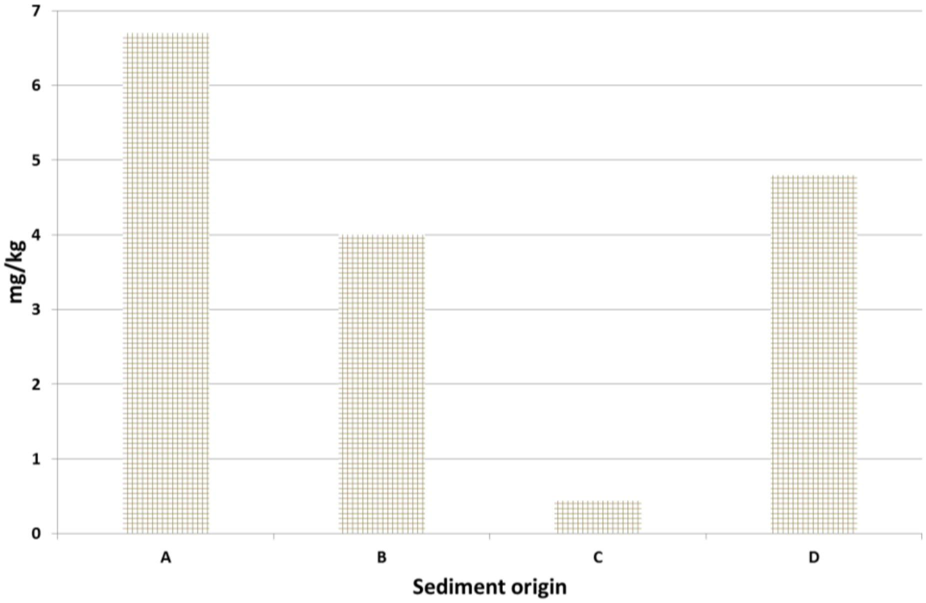

Figure 2 Comparison of ΣPAH in the different sediments. A = Fredhäll watershed, B = Södra Länken watershed, C = Asphalt pavement (abrasion test), D = Törnskogs tunnel watershed.

This would suggest that the road surfacing material is not a significant contributor to the PAH found in road runoff sediments. For Co, Cu, Pb and Zn, the concentrations in abrasion test sediment were 4-fold, 3-fold, 4-fold and 9-fold lower, respectively, than in the field sediments, suggesting that again, the asphalt road surfacing material is not the major contributor of these metals (Figs. 3-4, Tables 2-5). However, the asphalt road material could be a significant contributor of Cr and Ni to road runoff sediments. Previous studies have reported that Cr and Ni are present in traffic-related pollutants, but that low concentrations of Cr and Ni are associated with tyre and brake wear (McKenzie et al., 2009). Thus, other traffic-related activities such as asphalt road surfacing could be a significant source of Cr and Ni.

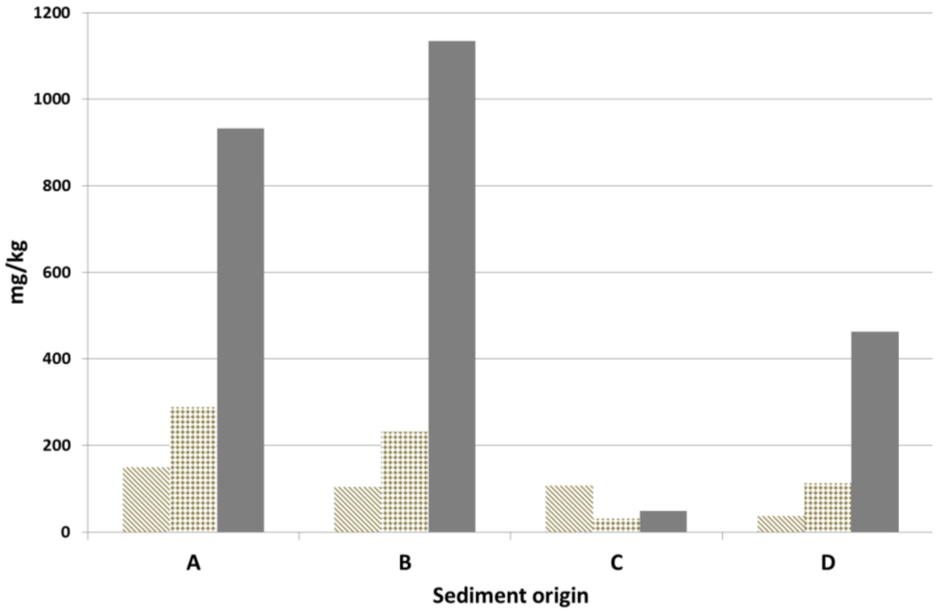

Figure 3 Comparison of Cr, Cu and Zn in the different sediments. Cross-hatched = Cr, Checked = Cu, Dark grey = Zn, A = Fredhäll watershed, B = Södra Länken watershed, C = Asphalt pavement (abrasion test), D = Törnskogs tunnel watershed.

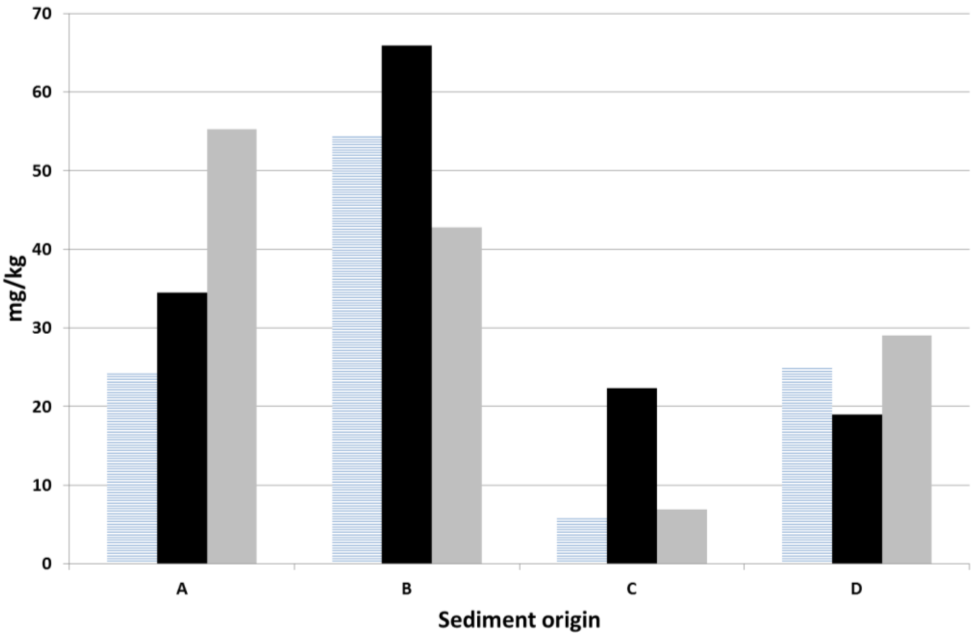

Figure 4 Comparison of Co, Ni and Pb in the different sediments. Blue bars = Co, Black bars = Ni, Light grey bars = Pb, A = Fredhäll watershed, B = Södra Länken watershed, C = Asphalt pavement (abrasion test), D = Törnskogs tunnel watershed.

The PAH concentrations in the sediments from the study sites were in the same range, suggesting that traffic is a major contributor to the PAH concentrations in road runoff.

The concentration of Zn in sediment from tunnel wash water was only half that in sediment from Fredhäll and Södra Länken (Fig. 3, Table 2-5). This suggests that there are additional contributors to Zn in addition to traffic and the traffic environment, one possibility being atmospheric deposition. The Pb concentrations in the sediment from the tunnel wash were 47% lower than those in the sediments from Södra Länken, which indicates that Pb pollution also has sources other than traffic (Fig. 4, Table 2-5).

In this study it was not possible to assess the capacity of the abraded particulate material from road surfacing material to transport other pollutants. Further studies would be of interest to assess this, in particular during winter when studded tires are used.

Table 2 Standard deviation (SD) and number of samples (n) for calculation of average concentrations of assessed parameters for the abrasion tests for samples A1, A2, B1, and B2.

	ΣPAH	Co	Cr	Cu	Ni	Pb	Zn
n	4	4	4	4	4	4	4
SD	0.266	2.281	28.40	8.60	2.83	1.197	17.89

Table 3 Standard deviation (SD) and number of samples (n) for calculation of average concentrations of assessed parameters from Fredhäll

	ΣPAH	Co	Cr	Cu	Ni	Pb	Zn
n	6	6	6	6	6	6	6
SD	1.62	3.18	46.8	31.0	3.25	4.10	90.0

Table 4 Standard deviation (SD) and number of samples (n) for calculation of average concentrations of assessed parameters from Södra Länken. * =*Detection limit 1.3 mg/kg to calculate average*

	ΣPAH	Co	Cr	Cu	Ni	Pb	Zn
n	31	35	35	35	35	35	35
SD	3.1*	10.8	11.5	43.4	6.7	3.3	222.1

Table 5 Standard deviation (SD) and number of samples (n) for calculation of average concentration of assessed parameters from the Törnskogstunnel

	ΣPAH	Co	Cr	Cu	Ni	Pb	Zn
n	6	6	6	6	6	6	6
SD	0.94	1.4	3.2	7.3	2.0	1.5	36.8

5 CONCLUSIONS

The concentrations of Cr and Ni found in asphalt road surfacing material indicate that it could be a major contributor to the Cr and Ni found in the sediments from road runoff in the three watersheds studied here. The PAH found in the runoff sediment evaluated probably had road traffic as a significant source. The Zn and Pb concentrations in runoff sediments were higher than could be explained by asphalt wear or traffic, and therefore must have additional sources. The results also showed that PAH, Co, Cu, Pb and Zn concentrations in road runoff sediments did not originate primarily from the asphalt road material.

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